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Supplementary Information

Monolithic Graded-Refractive-Index Glass-based Antireflective Coatings:

Broadband/Omnidirectional Light Harvesting and Self-Cleaning Characteristics

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Spectroscopic ellipsometry measurements and their interpretation:

Spectroscopic ellipsometry data were taken on a monolithic graded refractive index film using the two-modulator generalized ellipsometer (2-MGE)[31,32] at an angle of incidence of 65.01°. The raw data from the 2-MGE are 8 elements of the sample Mueller matrix, which can be reduced to 3 parameters, *N*, *S*, and *C* if the sample is isotropic and non-depolarizing. These parameters can be converted to a variety of other representations,

$$\rho = \frac{r_p}{r_s} = \tan(\psi) e^{i\Delta} = \frac{C + iS}{1 + N},$$

where r_p and r_s are the complex reflection coefficients for light polarized parallel and perpendicular to the plane of incidence, respectively. The quantities ψ and Δ are the traditional ellipsometry parameters. The quantity $\beta = (N^2 + S^2 + C^2)^{1/2}$ measures the depolarization, where $\beta = 1$ corresponds to no depolarization. The 2-MGE also measures parameters related to cross polarization, where pure *p*- (or *s*-) polarized light results in some *s*- (or *p*-) polarized light upon reflection; no significant cross polarization was observed with this sample.

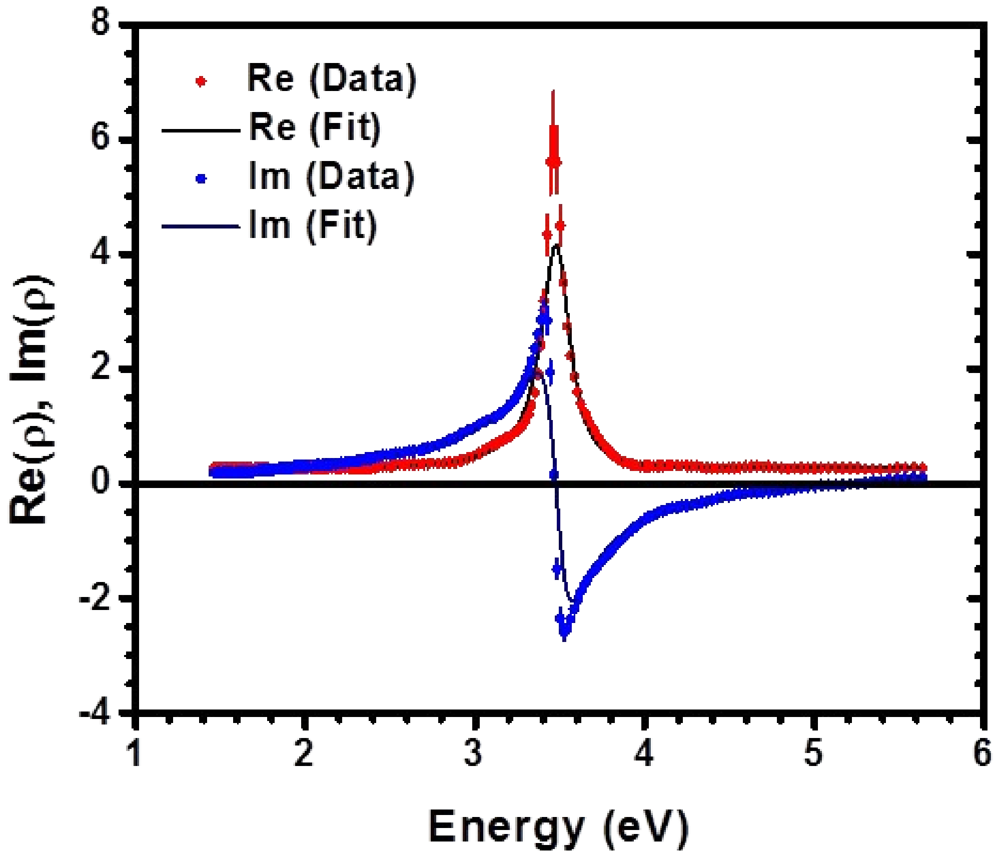
To gain an understanding of the optical structure of this film, we utilized the Levenberg-Marquardt fitting procedure, determining the reduced χ^2 as a figure of merit where the errors of the data are included (see references 31, 32). If $\chi^2 \leq 1$, then the model fits the data. The film structure considered consisted of 5 media: air/interface-1/film/interface-2/glass. The optical functions of interface-1 and interface-2 were determined using the Bruggeman effective medium approximation, consisting of 50% of the medium above and 50% of the medium below. The film and the glass optical functions were modeled using the Sellmeier approximation, where the dielectric function (or refractive index) is given by

$$\epsilon = n^2 = 1 + \frac{A\lambda^2}{\lambda^2 - \lambda_o^2}$$

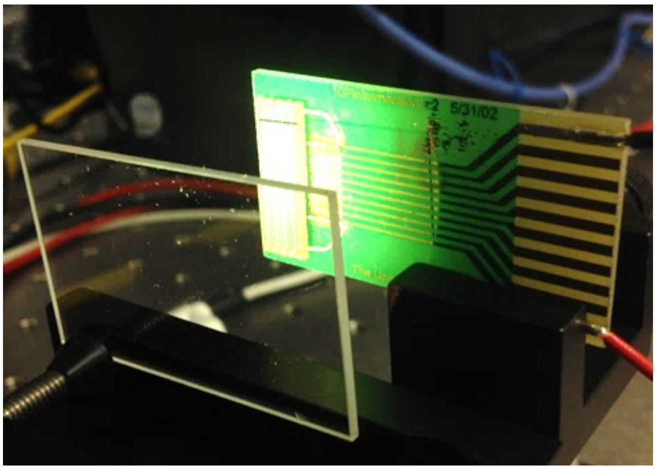
The wavelength is given by λ (in nm) and A is related to the oscillator strength. In a separate measurement of the glass, it was determined that A=1.165 and λ_o = 85.6 nm, resulting in n = 1.480 at 500 nm (slightly larger than fused silica).

The simplest model consisted only of air, the film and substrate, where the interfacial films were not taken into account. The resulting χ^2 = 23.0 for this fit, where fitted parameters were: 1) the film thickness = 123.1 nm, A = 0.2817, and λ_o = 169.4 nm (n =1.143 at 500 nm). If the interfacial layers are added to the model, then the resulting χ^2 = 11.8, and the 5 fitted parameters are given by: i) interface-1 thickness = 70.8 nm (n = 1.076); ii) film thickness = 67.9 nm; iii) film A = 0.3175; iv) film λ_o = 120.0 nm (film n = 1.154); v) interface-2 thickness = 36.5 nm (n = 1.307).

The best fit to the data was obtained using a 7-parameter fit, where we included a Gaussian distribution over the parameter A for the film. The fitted results presented here (and other intermediate fits not discussed here) show that the refractive index of the monolithic graded refractive index film is considerably less than the glass, and the refractive index is graded such that the surface refractive index is very close to that of air (n = 1.0).



Supplementary Figure S1. The real and imaginary parts of the spectroscopic ellipsometry data expressed in the complex reflectance ratio (ρ) representation. The solid lines are best fit to the data. Same samples as presented in Figures 5 and 6 are used for the measurements.



Supplementary Figure S2. Photograph of the experimental set-up to measure the photovoltaic performance of the solar cells (shown here is for Si). Coated and uncoated borosilicate substrates were placed in front of the cells to simulate the outer protective cover glass.

Supplementary Table ST1. Summary of the parameters obtained from fits to spectroscopic ellipsometry data for the same nanoporous glass coated borosilicate sample, where its optical properties are presented in Figures 5 and 6.

Parameter	Value	Error (±)
Interface 1 thickness (nm)	63.3	1.8
Film thickness (nm)	74.3	1.2
Film A	0.3066	0.0003
Film λ_o	130.3	1.5
Film A distribution	0.0477	0.0011
Interface 2 thickness (nm)	34.9	1.1
Substrate A	1.147	0.006

Video SV1: Al_2O_3 abrasion test: Impingement of Al_2O_3 particles onto a nanostructured antireflective surface at a velocity of 80 km h^{-1}.

Video SV2: Calibration of the **wind speed prior to** Al_2O_3 **abrasion test using a hot wire anemometer.**

Video SV3: Water droplets rolling from the sample surface after the Al_2O_3 abrasion test, demonstrating preservation of the water shedding characteristics of the nanostructured surface.